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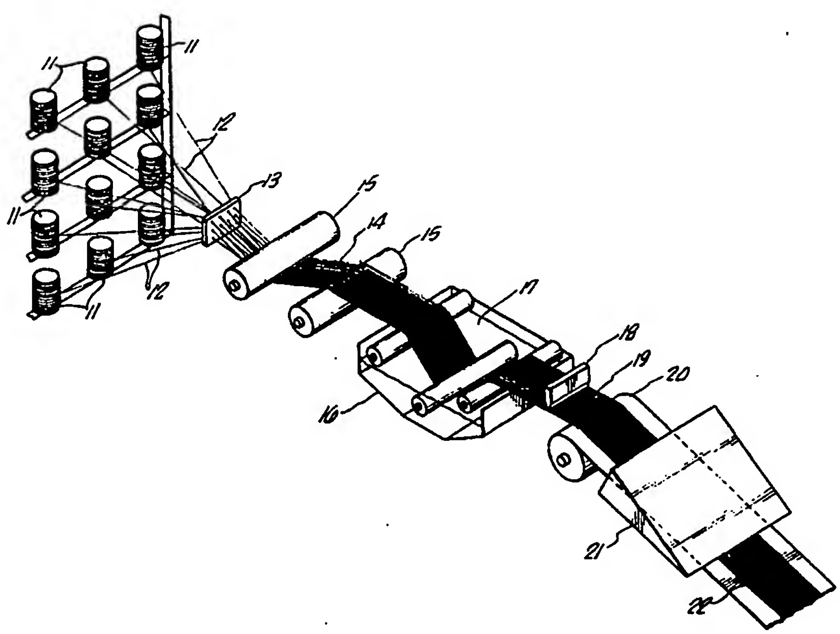
(54) Title: UNIDIRECTIONAL CARBON/PHENOLIC PREPREG MATERIAL AND METHOD OF MANUFACTURE



(57) Abstract

There are disclosed herein laminated materials often used as facing materials in aircraft interiors, and the use of phenolic resins and unidirectional carbon fibers (14) to form a prepreg, which ultimately comprises all of the layers of the laminated materials. Disclosed herein is a novel method for manufacturing such a prepreg and the product of that method. The final laminated material product formed has superior strength and impact resistance properties while exhibiting favorable smoke and burn characteristics.

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DESCRIPTION

UNIDIRECTIONAL CARBON/PHENOLIC PREPREG MATERIAL AND METHOD OF MANUFACTURE

FIELD OF THE INVENTION

This invention relates to laminated materials and the individual layers of which such materials are composed, and more particularly, to such materials for use in aircraft interiors.

In developing facing materials for use in aircraft interiors and other applications, desirable materials provide a high degree of strength, stiffness, and resistance to impact. These materials are commonly used in such applications as airplane interiors, cargo spaces and the like and are usually substantially planar, multi-layered materials which incorporate a synthetic resin. Ideally, in addition to offering optimal strength-weight properties, the laminated material should be easy to cut, and easy to handle during installation and repair. In many cases, the laminated assemblies contain a layer or layers of molding material which incorporates a synthetic resin. These resin-impregnated materials are frequently known by the term "prepregs." As used herein, and as used in the industry, this term refers to an intermediate product in the manufacturing process.

The invention disclosed herein relates to a resin-impregnated material or "prepreg," a multi-layered laminate assembly which incorporates the prepreg disclosed herein, and a method for making such a material. A preferred embodiment of the multi-layered laminate features superior strength, stiffness and impact resistance compared to similar materials presently known.

BACKGROUND OF THE INVENTION

Many different configurations and combinations of materials have been developed in an attempt to maximize

strength and stiffness, and impact resistance without adding additional weight. Typically, a multi-layered prepreg comprises a substantially planar core material overlaid on one or both surfaces with other materials which provide additional structural strength and may individually help to bind the layers together. A cross-section of such a material would reveal an inner core material with other layers applied to one or both planar surfaces.

The use of certain materials as the individual layers of a multi-layered prepreg assembly is well-known in the art. For example, use of woven glass cloth, fiberglass and carbon fibers is known and the general characteristics of each are familiar to those skilled in the art.

Most prepreg assemblies feature one or more layers containing a type of fiber such as glass or carbon. The properties of these layers and their methods of manufacture may vary. For comparison, common fiberglass material is a sheet comprising numerous tiny glass fibers in a random array to which a resin is added, and the resulting composition may be shaped into a desired form. Alternatively, individual fibers may be woven in a cloth-like fashion which may provide certain advantages in strength and allows easier handling and manufacture. Also known is the use of carbon fibers. To date, carbon fibers utilized in prepregs have been either woven or unidirectional. Unidirectional carbon fibers are not woven such that individual fibers run perpendicular to, and pass over and under one another; rather, all the individual fibers run parallel to one another and are commercially available in ribbon form wound onto spools.

Characteristic of prepregs is use of a synthetic resin which aids in holding the layers together and provides structural strength and stiffness. Years ago, phenolic resins were widely used, but the phenolic-based prepregs exhibited less than ideal properties because the phenolic resins were brittle and did not bind well to the materials

used as the core layer. With respect to the binding and brittleness properties, epoxy resins performed better than phenolic resins and offered advantages in the manufacturing process because epoxy resins could be used without first being dissolved in a solvent, which was necessary with the phenolic resins. Moreover, epoxy-based prepregs offered advantages in weight and strength and became widely used, largely phasing out the phenolic-based prepregs. The type of resin used also has a bearing on the type of fiber used. For example, when a solvent-based resin is used woven fibers are preferred because a solvent-based resin application system did not exist for use with unidirectional fibers. To date, the use of unidirectional fibers has been limited to non-solvent based, epoxy resin application systems.

Unfortunately, prepreg materials used in aircraft interiors, occasionally and often tragically, came into contact with flames or high temperatures. In such cases, the epoxy resins were found to have dangerous ignition and burn characteristics and to emit toxic smoke in certain high temperature conditions. When the dangerous properties of epoxy resins became known, the United States Federal Aviation Administration promulgated regulations, the effect of which was to promote a phasing out of the epoxy resins notwithstanding the known advantages. Phenolic resins again became widely used despite drawbacks inherent in a manufacturing system utilizing a solvent. At this time, woven fibers became the industry standard rather than unidirectional fibers because woven fibers could be easily handled when used with solvent-based resin solutions. Accordingly, in most processes used today, a fabric of woven fibers is impregnated with a phenolic resin/solvent solution and dried, thus forming the prepreg.

SUMMARY OF THE INVENTION

The present invention enables the use of unidirectional carbon fibers with phenolic resin/solvent solutions. A manufacturing process is described herein which produces sheets of phenolic-resin impregnated unidirectional carbon fibers which are ultimately used in forming a laminated assembly. Testing data has revealed that laminated assemblies which incorporate these prepregs have superior strength, stiffness and impact resistance properties while maintaining favorable smoke and burn characteristics.

Therefore, in the present state of the art, most aircraft interior prepregs are comprised of woven fiberglass or woven carbon fibers which are coated with a solution of resin and laminated to both sides of a paper honeycomb core. To modify a particular prepreg design as a function of the allowable weight and desired strength, varying thicknesses of the core and different types of fiberglass or carbon fibers may be used. Additionally different quantities, types and application processes of resin can be used.

The amount of carbon present in an individual type of prepreg may be described by the "Areal weight" of the prepreg which measures the grams of carbon in a square meter of the final prepreg. The commercially available Amoco EXAS-33-500 epoxy-based prepreg has an Areal weight of about 150 g/m^2 and a laminated assembly featuring this prepreg has an impact resistance of approximately sixty inch-pounds.

For comparison, an embodiment of the present invention with an Areal weight of 150 g/m^2 has an impact resistance of up to 140 inch-pounds. Similar impact-resistance properties have been achieved with embodiment of the present invention with an Areal weight as low as 120 g/m^2.

In terms of gross weight of the resulting prepreg, model L-525, 1583 style prepreg manufactured by J.D. Lincoln, Inc. weighs 1.6 times as much per area as that made according to the present invention and will withstand

an impact of eighty-five inch-pounds. Another prepreg manufactured by Fibrerite, Inc., and used in the McDonnell-Douglas MD-80, will withstand an impact of ninety inch-pounds but weighs twice as much as that of the present invention.

It is an object of the present invention to provide an improved prepreg material.

It is an object of the present invention to provide an improved multi-layered laminate assembly.

It is also an object of the present invention to provide an improved high-strength, low-weight, multi-layered material that does not possess dangerous toxic fume or combustion characteristics.

It is a further object of the present invention to provide a new process for making an improved prepreg material and an improved multi-layered laminate assembly.

DETAILED DESCRIPTION OF THE INVENTION

The invention will be more readily understood by reference to the accompanying drawings, in which,

FIGURE 1 illustrates a preferred embodiment of the present invention showing the individual layers of the final laminated assembly separated from each other to reveal the relationship of the individual layers of which the final prepreg is comprised.

FIGURE 2 illustrates an apparatus which may be used in the manufacturing process of an individual sheet of resin-impregnated unidirectional carbon fibers.

FIGURE 3 illustrates a preferred embodiment of the present invention showing a final laminated assembly with a partial cut-away of each individual layer to show one possible orientation of the individual layers.

The concepts of the present, and an exemplary embodiment thereof, will best be understood through a description of the individual layers and the manufacturing process of the individual layers and the final product.

Referring to FIGURE 1, the inner core material may comprise a number of materials which are low in cost and light in weight. A preferred embodiment of the present invention utilizes a core 1 composed of an aramid paper pre-formed through a conventional process into a honeycomb pattern and dipped in phenolic resin.

Another layer, often used in other prepreg configurations sold in the industry, is an adhesive film layer 2 sold under the name "Bond-Aid" (product number L-310) by J.D. Lincoln, Inc., Costa Mesa, California, which was developed by this inventor, and is used in the preferred embodiment of the present invention. The film contains a phenolic resin and, although its structural properties are unknown, the adhesive film layers facilitate binding the inner core 1 to the outer layers.

Sheets of unidirectional carbon fibers 3 and 3' are prepared by a novel process in which the fibers are immersed in a resin bath after being arranged into a sheet of uniform thickness. A novel manufacturing process for preparing such resin-impregnated fibers is disclosed herein, although impregnation of fibers with a synthetic resin is a known procedure that may be accomplished in a variety of ways. Layers 3 and 3' are two layers of resin-impregnated unidirectional carbon fibers which are identical in manufacture and are oriented substantially perpendicular to one another such that the individual carbon fibers in the sheet are in a 0°/90° orientation. The 0°/90° orientation shown is a preferred embodiment although other orientations may be desireable depending on the strength and impact resistance required and the weight limitations of a particular application. Layers 4 are composed of a glass scrim cloth which is attached to the layers of carbon fibers 3 and 3'. Ultimately, this assembly is cured in a laminating press to fuse the layers together for use as the final prepreg product.

As noted above, a novel process for manufacturing continuous sheets of resin-impregnated unidirectional

carbon fibers is disclosed. Referring to FIGURE 2, one preferred method is to draw strands or "ribbons" of Toray T-700 carbon fibers **12** from several spools **11**. The number and orientation of the individual spools is for illustrative purposes only; in practice the number of spools **11** may be far greater, and the spools **11** could be placed in many suitable arrangements. The number of spools will vary as a function of the width and thickness of the spooled carbon fiber strands and the weight of carbon desired in the prepreg. After being removed from the spools **11**, the strands of carbon fibers **12** are passed through a guide **13** or series of guides which are oriented to align and assemble the strands to form a uniform sheet **14** of the desired width and thickness. Increasing the thicknesses of the sheet of the fibers thus assembled requires only the use of more spools of carbon fiber. Further uniformity in thickness can be achieved by passing the strands of carbon fiber under and over a series of cylindrical bars **15** such that the bars slightly deflect the path of the strands. The two cylindrical bars **15** shown in FIGURE 2 are meant to be representative of one or more of such apparatus as needed to create a continuous sheet of carbon fibers of uniform thickness. At this stage, the strands of carbon fibers should form a sheet of substantially uniform thickness without gaps or holes in the sheet so that after being immersed in resin the fiber sheet of carbon fibers is continuous and uniform.

Rather than pressing a previously manufactured film layer of resin onto a layer of whatever type and configuration of fibers are being used, the present invention describes a superior method of applying the resin. Again referring to FIGURE 2, impregnating fibers with resin through the use of a resins dissolved in a solvent in a bath **16** rather than a pre-formed resin film reduces the viscosity of the resin which is believed to increase the degree to which the resin impregnates the fibers. It is further believed that this method increases the bonding of

the resins to the fiber substrate. Once the fibers are oriented as above, the resulting continuous sheet is then passed through a resin bath **16** to coat and impregnate the carbon fibers with phenolic resin. The phenolic-resin bath solution **17** contains liquid resin dissolved in a solvent. In a preferred embodiment, the resin is the commercially available phenolic resin Chem-Bond 6010. The phenolic-resin solution, as commercially supplied, contains 60% phenolic resin solids dissolved in methanol. To formulate the resin-bath solution **17**, ethanol is added to bring the specific gravity of the resin-bath solution to approximately 0.94 g/ml.

Continuing to refer to FIGURE 2, when the sheet of carbon fibers emerges from the resin bath, the resulting resin-impregnated continuous sheet of unidirectional carbon fibers is drawn underneath an edge **18** that removes excess resin from the sheet of fibers. This continuous sheet of unidirectional carbon fibers **19** is applied to a roll of silicone-coated paper **20** and passed through an oven **21** for six minutes with an ambient air temperature of approximately 210°F ± 10°. The resulting sheet of resin-impregnated unidirectional carbon fibers **22**, which is incorporated into the final prepreg, contains approximately 36-44% phenolic resin solids.

Once each layer is prepared, formation of an intermediate prepreg product is performed. A first layer of the unidirectional carbon fibers is prepared in a desired shape and size by laying out a series of the sheets of resin-impregnated carbon fibers with the treated-paper backing. Additional layers of resin-impregnated carbon fibers are applied to the first layer. Taking advantage of the treated paper backing, additional layers are applied so that the carbon fibers of each layer face and contact one another and so that the treated paper backing can be peeled off in preparation for applying another layer of carbon fibers or a different component of the final laminated assembly.

In a preferred embodiment, a second layer of unidirectional carbon fibers is laid on top of, and substantially perpendicular to, the first layer. These two layers are massaged by rolling metal rollers across the flat surface of the combined layers to create a smooth layer and to encourage bonding between the layers. This assembly is termed a "0°/90° unidirectional carbon fiber prepreg." The treated paper is removed from one side of this assembly. To the side with the paper removed is added a layer of type 108 glass scrim cloth which covers one entire surface of the resulting prepreg and thus becomes an exterior surface of the final prepreg. This layer eases handling of the resulting prepreg and prevents fraying of the prepreg and other underlying material if the prepreg is cut to fit its intended use.

In one preferred embodiment, another assembly comprising two layers of fibers **22** and one layer of glass scrim cloth **4**, identical in size and manufacture to the one described immediately above, is prepared. Next, a layer of adhesive film **2** is attached to both sides of a honeycomb paper core material **1** of the desired shape. Finally, the remaining paper backing is removed from the layers of carbon fibers and glass cloth, and the carbon fiber layer is placed most proximate to the core material so that a layer of carbon fibers directly contacts the adhesive film layer. Accordingly, in this embodiment of this invention the glass cloth layer constitutes both exterior layers of the final laminated assembly.

Once all layers are combined, the final laminated assembly is placed in a laminating press and cured for about ten minutes to two hours at a temperature of approximately 260-350°F at approximately 25-200 psi, preferably at about fifty psi.

Typically, the resulting laminated assembly is 10-12mm thick, although the thickness can be varied depending on the allowable weight and desired strength.

In another preferred embodiment, the unidirectional carbon fiber layers, the adhesive film layers and the layer of glass scrim cloth are combined without the inner core material. A 0°/90° unidirectional carbon fiber prepreg is prepared as described above. A layer of the glass scrim cloth is attached to one surface and the adhesive film layer is attached to the other surface. This assembly is bound together by placing it in a laminating press at approximately 5 psi and 125-175°F for ten minutes to two hours. This step may be performed under vacuum although the quality of the final prepreg does not appear to be affected by the vacuum environment. Therefore, in this embodiment the layers of the prepreg are combined without the inner core material so that the essential elements of the prepreg may be commercially available without being bound to a particular core material.

It will be apparent to one skilled in the art that the invention and embodiments disclosed herein are susceptible to various modifications and alterations. Accordingly, the present invention should not be limited to only the particular embodiments described.

WHAT IS CLAIMED IS:

1. A material comprising unidirectional carbon fibers impregnated with a solvent-based synthetic resin.

2. The material of claim 1 wherein the synthetic resin is a phenolic resin.

3. A material comprising layers of unidirectional carbon fibers impregnated with a synthetic resin and oriented substantially in a 0°/90° configuration.

4. The material of claim 3 wherein the resin is phenolic resin.

5. A multi-layered material comprising a flat two-sided core material having a layer of adhesive film, unidirectional carbon fibers impregnated with a synthetic resin and oriented substantially in a 0°/90° configuration, and glass scrim cloth.

6. The material of claim 5 wherein the core material is aramid paper pre-formed into a honeycomb structure and dipped in phenolic resin.

7. The material of claim 5 wherein the adhesive film layer contacts the core material.

8. The material of claim 5 wherein the material comprises a flat two-sided core, a layer of adhesive film on each side of the core, said carbon fibers form layers secured to each side of the core by the adhesive film, and the glass scrim cloth comprises two layers of glass scrim cloth on the respective carbon fiber layers as the exterior layers of the material.

9. The material of claim 5 wherein the unidirectional carbon fibers are impregnated with a phenolic resin.

10. The material of claim 5 wherein the adhesive film layer comprises soluble nylon copolymer and a phenolic resin.

11. A multi-layered material comprising an inner honeycomb aramid paper core, a layer of adhesive film bound to both surfaces of the inner core, a layer of unidirectional carbon fibers impregnated with a phenolic resin oriented substantially in a 0/90° configuration ...

attached to the adhesive film layers, and a layer of glass scrim cloth comprising the exterior surface.

12. A process for manufacturing a material which comprises impregnating unidirectional carbon fibers with a synthetic resin by passing an assembly of fibers through a bath of synthetic resin dissolved in an alcohol solvent.

13. The process of claim 12 wherein the synthetic resin is a phenolic resin.

14. The process of claim 12 wherein the resulting sheet of resin-impregnated carbon fibers is cured by passing it through an oven.

15. A process for manufacturing a material which comprises impregnating unidirectional carbon fibers with a synthetic resin by passing an assembly of fibers through a bath of synthetic resin dissolved in an alcohol solvent and curing the resulting sheet by passing it through an oven;

forming a multi-layer sheet of unidirectional carbon fibers in a 0°/90° orientation by pressing together sheets of the resin-impregnated unidirectional carbon fibers having been laid substantially perpendicular to one another;

attaching a sheet of glass cloth to one surface of the 0°/90° oriented unidirectional carbon fibers;

attaching an adhesive film layer to both sides of an inner core material;

attaching a layer of carbon fibers and glass cloth to each side of the inner core and adhesive film such that the glass cloth layer is the outermost layer; and

curing the resulting multi-layered material by laminating these layers in a press at a temperature from about 260° for two hours to about 350°F for about ten minutes.

16. The process of claim 15 wherein the synthetic resin is a phenolic resin.

17. A process for manufacturing a material which comprises impregnating unidirectional carbon fibers with a synthetic resin by passing an assembly of fibers through a bath of synthetic resin dissolved in an alcohol solvent and drying the resulting sheet by passing it through an oven with an ambient air temperature of about 210°F ± 10°F;

forming a multi-layer sheet of unidirectional carbon fibers in a 0°/90° orientation by pressing together sheets of the resin-impregnated unidirectional carbon fibers having been laid substantially perpendicular to one another;

attaching a sheet of glass cloth to one surface of the 0°/90° oriented unidirectional carbon fibers;

attaching an adhesive film layer to the surface of the 0°/90° oriented unidirectional carbon fibers not covered with glass scrim cloth;

pressing the assembled layers in a laminating press at approximately 5 psi and about 125° - 175°F for ten minutes to two hours.

18. The process of claim 17 wherein the synthetic resin is a phenolic resin.

19. The process of claim 17 wherein the assembled layers are pressed together in a laminating press under vacuum conditions.

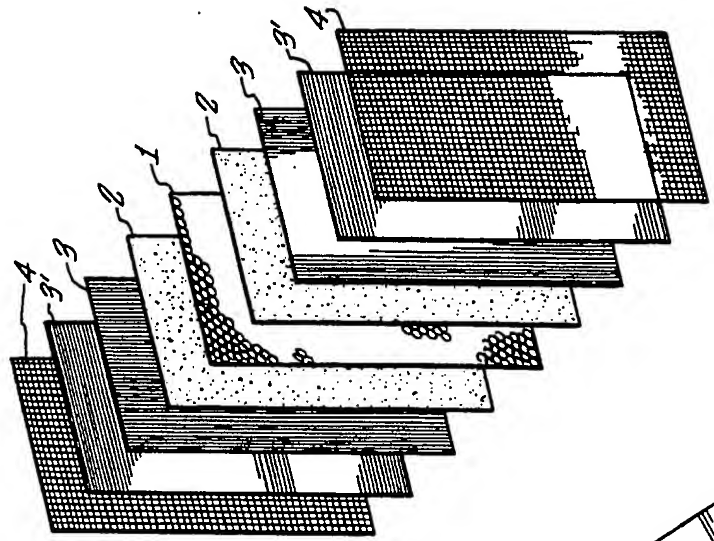

FIG. 1.

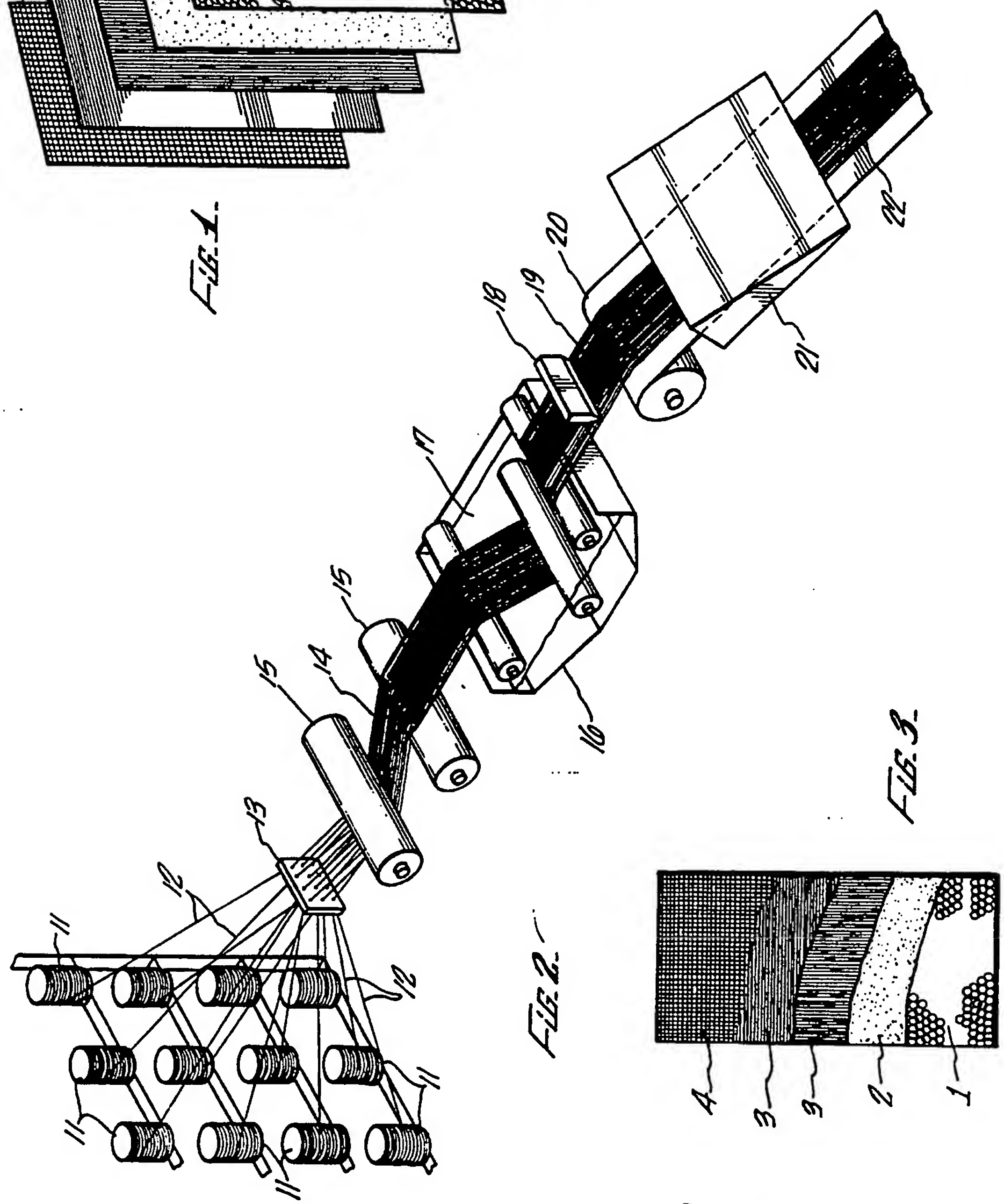

FIG. 2.

FIG. 3.

INTERNATIONAL SEARCH REPORT

International Application No. PCT/US92/02797

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) [6]

According to International Patent Classification (IPC) or to both National Classification and IPC

IPC (5): B32B 3/12

U.S. CL. 156/292; 428/116

II. FIELDS SEARCHED

Minimum Documentation Searched [7]

Classification System	Classification Symbols
U.S.	156/292; 428/116

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched [8]

III. DOCUMENTS CONSIDERED TO BE RELEVANT [9]

Category *	Citation of Document, [11] with indication, where appropriate, of the relevant passages [12]	Relevant to Claim No. [13]
Y	US, A, 4,622,091 (LETTERMAN) 11 NOVEMBER 1986. See entire document.	1-19
Y	US, A, 4,900,048 (DERUJINSKY) 13 FEBRUARY 1990. See entire document.	1-19
Y	US, A, 4,917,742 (WATANABE et al) 17 APRIL 1990. See entire document.	1-19

* Special categories of cited documents: [10]

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IV. CERTIFICATION

Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report
30 June 1992	13 JUL 1992
International Searching Authority	**Signature of Authorized Officer**
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